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Positron lifetime studies of 100-MeV oxygen irradiated Pb-doped Bi-2223 superconductors

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Abstract

Positron lifetime studies have been carried out for unirradiated and 100-MeV oxygen ion irradiated Pb-doped Bi-2223 superconductors. The analysis of positron lifetime spectra revealed three lifetime components: a short lifetime, $\tau_1 = 153$ –196 ps; an intermediate lifetime, $\tau_2 = 269$ –339 ps; and a long lifetime, $\tau_3 = 616$ –812 ps. A decrease in all the lifetime components, τ_1 , τ_2 and τ_3 with an increase of the relative intensities of I_2 and I_3 has been observed with increasing fluence. The positron lifetime results obtained are contradictory to the expected trend for the irradiated samples. This variation has been attributed to the formation of cationic clusters and their segregation at the grain boundaries; and probably in between the Bi–O planes too, where the positron density distribution is a maximum. This is highly possible since the metal atoms have a fair chance to get displaced from their original positions as a result of swift heavy ion irradiation. The growth of the Bi-2212 phase with irradiation supports the positron lifetime results. © 2000 Published by Elsevier Science Ltd. All rights reserved.

Keywords: A. High- T_c superconductors; D. Electronic transport; D. Radiation effects; E. Positron spectroscopies

1. Introduction

After the discovery of high temperature superconductors, in 1986, by Bednorz and Muller [1], several efforts have been made to understand the mechanism of superconductivity in them. Several experimental probes have been employed to obtain valuable information of the various aspects of these superconductors, viz. structural, electronic transport, microstructure, bandstructure, etc. [2–4]. Positron annihilation technique is a powerful non-destructive tool, which provides an insight to the electron density, nature and concentration of defects and electron momentum distribution from the positron annihilation rate, angular distribution of annihilation photons and Doppler broadening of the emitted gamma photons [5–7]. A serious problem in understanding the mechanism of superconductivity is the difficulty in the availability of defect free samples. However,

it is known that for any application of superconductors, defects are not only unavoidable but also very much desirable [8,9]. Swift heavy ion irradiation (SHI) [10] is one of the widely used methods to produce controlled defects inside these materials. In this communication, positron lifetime technique has been employed to probe the defects generated in bulk Bi(Pb)-2223 superconductor as a result of irradiating them with swift heavy ion of oxygen with 100 MeV energy.

Most of the positron annihilation studies done so far on high temperature superconductors [11–15] have concentrated on the nature of change in the lifetime of the superconducting and the normal state as the superconductor is cooled down through the superconducting transition temperature. Many ascribe the changes to the dimerisation of oxygen atoms wherein the oxygen–oxygen interaction is mediated by Cu ions, while several others ascribe it to the local electron transfer from CuO_2 to Bi–O layers resulting in the generation of holes in the CuO_2 plane, and a concomitant increase in the electron density in the Bi–O planes at the onset of superconductivity. A recent work by Sen et al. on Bi-2212 and Bi-2223 superconductors shows that

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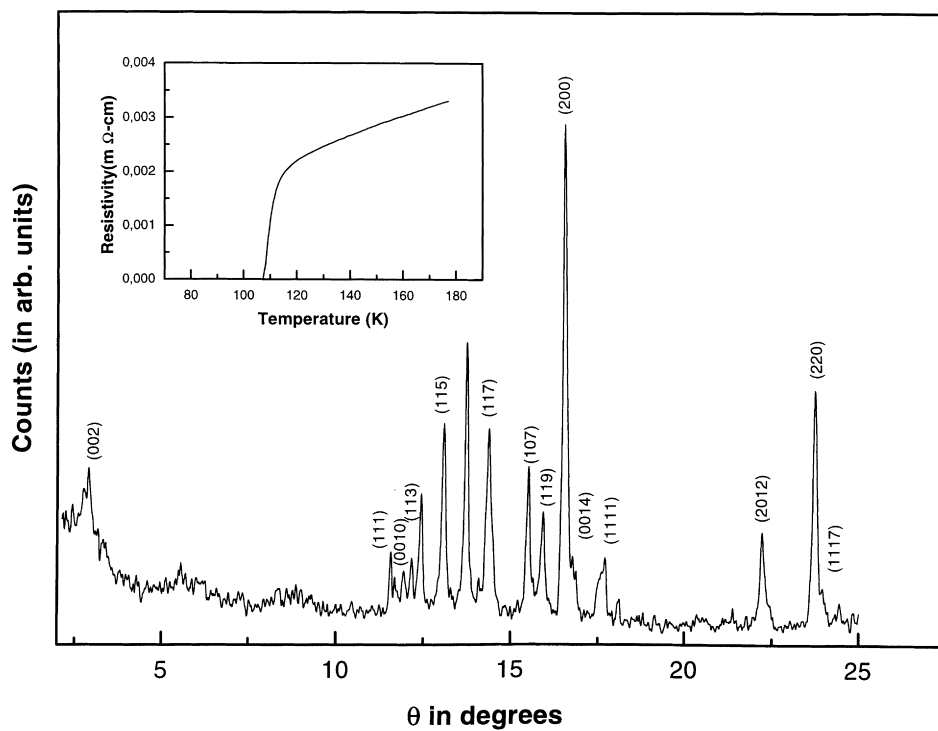


Fig. 1. X-ray diffraction pattern of unirradiated Bi(Pb)-2223. Inset shows the ρ – T plot of unirradiated Bi(Pb)-2223.

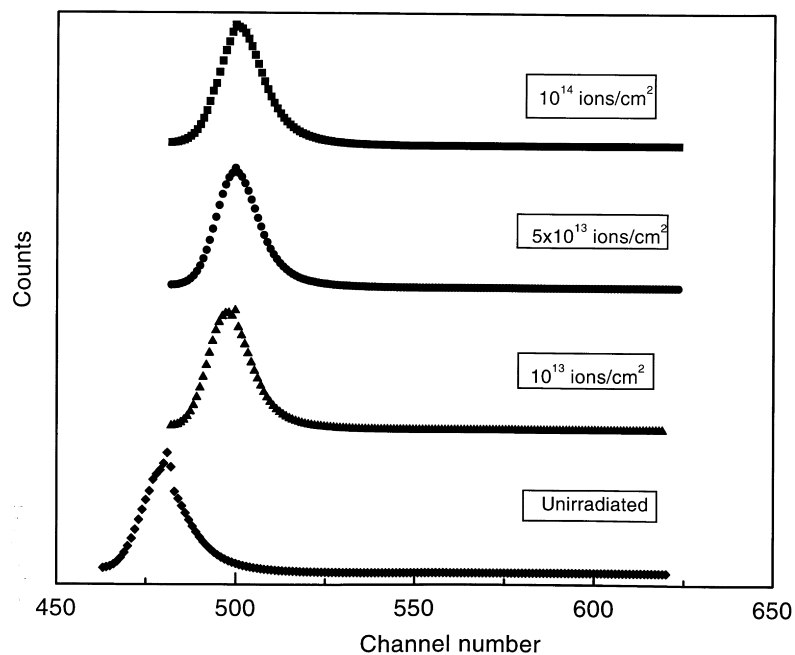


Fig. 2. Positron lifetime spectra of unirradiated and irradiated Bi(Pb)-2223.

Table 1
Positron lifetime results for unirradiated and irradiated Bi(Pb)-2223 superconductors

Sample	Positron lifetime parameters					
	τ_1 (± 2) (ps)	τ_2 (± 5) (ps)	τ_3 (± 70) (ps)	I_1 (%)	I_2 (± 2) (%)	I_3 (± 0.5) (%)
Bi(Pb)-2223 unirradiated	196	339	882	74	25	<1
Irradiated: 10^{13} ions/cm ²	147	296	797	61	37	<2
Irradiated: 5×10^{13} ions/cm ²	141	277	642	57	40	≈ 3
Irradiated: 10^{14} ions/cm ²	153	269	616	45	51	≈ 4

divacancies and monovacancies are the predominant trapping sites of positrons in proton irradiated samples of Bi-2212 and Bi-2223 [16].

2. Experimental details

The high purity pellets of Pb-doped Bi-2223 samples were prepared by the solid-state reaction route involving

repeated calcination in air at temperatures of 790, 830 and 840°C for a total span of 30 h, and sintering after pelletising in the temperature range of 855–858°C for a total period of 150 h. X-ray diffraction at room temperature and resistivity versus temperature plot (as shown in Fig. 1) established the phase purity of an unirradiated sample. 100-MeV oxygen irradiation was carried out using the 15 UD Pelletron accelerator at Nuclear Science Centre, New Delhi. The beam

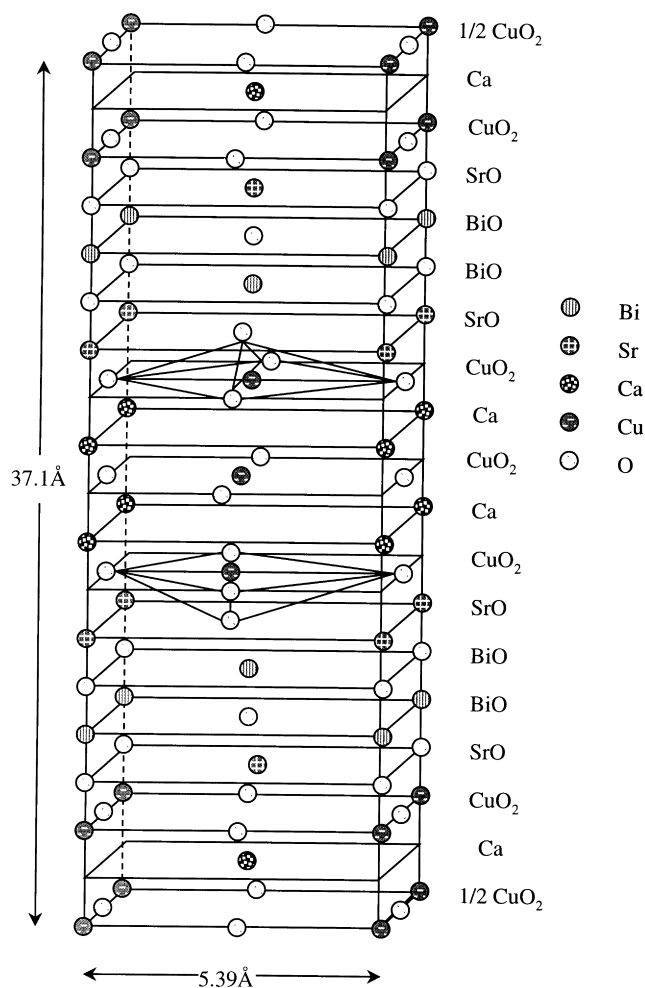


Fig. 3. Crystal structure of Bi-2223.

current used was 10 pA ($\sim 6.25 \times 10^{10}$ particles/cm²/s). The beam was scanned across the sample so as to uniformly irradiate it. The samples were irradiated in a secondary electron suppressed geometry in a vacuum of 2×10^{-6} mbar/cm at fluences of 10^{13} , 5×10^{13} ions/cm², and 10^{14} ions/cm². The positron lifetime measurements on these samples were made at room temperature. The radioactive source Na²², with an activity of 10 μ Ci was used for the positron lifetime measurements. The source was sandwiched in between the two identical pellets of the same specimen. The lifetime was measured by the standard method of fast–slow coincidence [17]. The time resolution of the set-up obtained from the prompt Co⁶⁰ spectrum was 280 ps. The analysis of the lifetime spectra was done using the program POSITRONFIT [18]. The uncertainty caused by single Gaussian representation of the resolution function was reduced by excluding the peak region of the spectrum from the analysis, i.e. by starting from the ($T_0 + 4$ th) channel.

3. Results and discussion

Positron lifetime spectra for the Pb-doped Bi-2223 superconductor are shown in Fig. 2 for the unirradiated case as well as for the irradiated cases. These spectra could be fitted with three lifetime components: a short lifetime component, τ_1 varying from 196 ps in the case of the unirradiated sample to 153 ps for the highest fluence; an intermediate lifetime τ_2 varying from 339 to 269 ps; and a relatively larger third lifetime τ_3 varying from 882 ps in the unirradiated case to 616 ps for the highest irradiated sample. The positron lifetime values τ_1 , τ_2 and τ_3 and their corresponding intensities I_1 , I_2 and I_3 are listed in Table 1. The lifetime component τ_1 corresponds to the annihilation with the bulk lattice, τ_2 corresponds to the trapped state annihilation and τ_3 corresponds to the annihilation by formation of *ortho*-positronium (*o*-Ps), subsequently going to the *para*-positronium (*p*-Ps) state. Considering the low values of I_3 , the two-state trapping mode of annihilation can be assumed to be valid for the present case.

The value of τ_1 , quite consistent with the previous investigations on this system [15,16], corresponds to the lifetime of delocalised positrons and free annihilation within grains. In this system, the positron density distribution (PDD) in the Bloch state is believed to be mainly confined between the Bi–O layers and is slightly overlapped with superconducting CuO₂ layers [15]. The τ_1 value is found to decrease with an increase of ion fluence, which signifies a higher PDD in the Bloch state in these cases.

The value of τ_2 , in general corresponds to the lifetime of positrons trapped by larger defects or defect clusters in the crystal lattice. Defects caused by irradiation in Bi-2223 should act as efficient positron traps leading to an increase of lifetime by decreasing the probability of annihilation. With an increase of ion fluence, the defect size should either increase or remain constant. Subsequently, τ_2 should

increase or remain constant. However, the results obtained on irradiation are quite contradictory. The value of τ_2 decreases monotonically with an increase of the ion fluence, while the concentration of the defects as indicated by I_2 , increases with an increase of the ion fluence. SHI irradiation leads to the displacement of cations from their atomic sites. Such a displacement of metallic atoms usually occurs in proportion to their scattering cross-section. A gradual build-up of a second phase of Bi-2212 is also evident from the XRD patterns of the irradiated samples [19]. SHI irradiation on these superconductors causes a decrease of the relative intensities of the (117) and (119) planes [20]. This decrease of intensity arising from the coherent scattering of the atomic planes is related to the displacement of certain cations from them. Displacement of atoms from their original atomic site is also evident from STM studies of these irradiated samples [20,21]. Looking at the unit cell of the Bi-2223 system (Fig. 3), one can visualise that the possible atoms residing at the (117) and (119) planes are Bi, Ca and Cu. The growth of the lower phase of Bi-2212 is also seen with increasing fluence. This is possible only if one Ca and one Cu atom is removed from the Bi-2223 lattice. It can be inferred that these atoms segregate and form cationic clusters, probably at the grain boundaries and in between the Bi–O layers where the positron density distribution is a maximum [14]. The increased metallicity with increasing fluence can also be confirmed if one looks at the slope of the ρ – T curves of the irradiated samples at temperatures close to room temperature [19]. The anomalous decrease in the τ_2 values with an increase of irradiation fluence can thus be explained by the quick annihilation of the positrons on encountering such electron density rich centres in the samples. HRTEM studies on these samples can be carried out to locate the sites of such atomic displacements as well as to establish the exact size of these metallic clusters.

The third larger component which has been fitted to get correct values of τ_1 and τ_2 can very well be attributed to *o*-Ps formation in voids and intergranular spaces and the subsequent conversion of *o*-Ps to *p*-Ps in the conducting oxide samples used here. The decrease of the τ_3 value also suggest the enhanced metallicity set up in the Bi-2223 samples with an increase of the ion fluence.

4. Conclusions

In summary, the positron lifetime spectra of 100-MeV oxygen ion irradiated Pb-doped Bi-2223 superconductors have been fitted with three lifetime components. A simultaneous decrease of the τ_1 , τ_2 and τ_3 values with increasing ion fluence has been observed. This is a definite indication of the development of enhanced metallicity in the samples at room temperature as a result of irradiation. The decrease in the lifetime components is ascribed to the knock out and clustering of certain cations at the grain boundaries and in the region between the Bi–O layers.

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